SYNOPSIS

Exploring Metastable Energy States through Engineered Phase Competition in Manganite Systems

SUBMITTED BY

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1. Title: Exploring Metastable Energy States through Engineered Phase Competition in Manganite Systems

2. Introduction and Background

Manganites $R_{1-x}A_xMnO_3$ exhibit a rich variety of electronic and magnetic phenomena arising from the competition among double exchange, superexchange, Jahn–Teller lattice coupling, and quenched disorder [1,2]. These competing interactions give rise to metastable energy states manifested through hysteresis, slow relaxation, memory effects, and electronic phase separation (EPS) [1,3,4]. EPS, in which ferromagnetic metallic and antiferromagnetic charge-ordered insulating regions coexist, plays a decisive role in transport and magnetic responses, particularly near metal–insulator transitions [1,5,6]. Additionally, geometrically frustrated manganites—such as hexagonal $RMnO_3$ and layered compounds—host non-collinear or glass-like magnetic states due to constraints on long-range ordering imposed by lattice geometry [7,8]. Understanding these metastable and frustrated regimes requires a microscopic framework capable of simultaneously treating electronic correlations, lattice distortions, and competing magnetic interactions [1,9,10].

Although substantial progress has been made [2], key gaps remain in the microscopic understanding of metastability in manganites. Experiments show that geometric engineering, such as antidot arrays and confined thin-film geometries, can dramatically modify phase coexistence and shift the metal–insulator transition [5,6]. However, there is no comprehensive theoretical framework that explains how symmetry breaking, nanoscale confinement, or artificial patterning alter free-energy landscapes and phase stability in AFM–CO systems [9,10,11]. Likewise, the combined influence of charge density, dimensionality, disorder, and engineered lattice modifications on EPS and CMR-like behavior has not been systematically addressed [9,10,11]. In frustrated lattices, non-collinear and glassy states are known to emerge [7,8], but the effect of artificial structuring on frustrated energy landscapes and metastable configurations remains unexplored.

The spin–fermion model, solved using Monte Carlo simulations, provides an ideal platform for addressing these questions because it naturally incorporates itinerant electrons, localized spins, electron–lattice coupling, and disorder [12,13,14]. It successfully reproduces EPS, frustration-induced magnetism, slow dynamics, and metastability [1,3,4], making it well suited for probing how artificial geometric features and electronic tuning reshape phase competition. The present study is therefore motivated by the need for a unified microscopic investigation that explains how metastable states originate and how they can be deliberately manipulated using antidot arrays, charge-density variation, thickness control, and frustrated lattice geometries. Limited experimental measurements will serve as qualitative validation, ensuring that the theoretical predictions are physically grounded [2,5,6].

3. Objectives

To perform a comprehensive theoretical investigation of metastable energy states in manganites using spin–fermion Monte Carlo simulations [12,13,14], with an emphasis on manipulating

and understanding electronic phase separation and frustration-driven metastability through engineered structures and parameter variation. The specific objectives are:

I. **Manipulating EPS via ordered antidot arrays:** Model AFM–CO perovskite systems in which ordered arrays of antidots (of varying shapes, sizes, and periodicities) are embedded [5,15]. Study how antidot geometry and edge-induced symmetry breaking influence nucleation, growth, stabilization, and melting of ferromagnetic metallic (FMM) domains, and quantify the resultant shifts in metal–insulator transition temperature and transport properties [9,10,11].

II. **Charge-density dependence in AFM–CO models:** Investigate AFM–CO model systems at different charge carrier densities [11,16] to map how electronic filling affects the balance between COI and FMM phases, the degree of EPS, metastability, and the magnitude/temperature-location of resistivity peaks associated with CMR-like behavior [1,9,10].

III. **Thickness-dependent thin-film effects:** Simulate thin-film geometries of varying effective thickness to study dimensional crossover effects on EPS and metastability [5,6]. Examine how film thickness modifies domain morphology (including FMM rings near antidots), strain-like boundary effects, and the energetics of phase nucleation in confined geometries [6,15].

IV. **Artificial-structure control of metastability in frustrated lattices:** Explore geometrically frustrated manganite lattices (triangular, kagome-like, FCC-derived motifs) with artificial patterning (antidots, patterned vacancies, or imposed boundary conditions) [7,8,17] to reveal new metastable magnetic or electronic states, changes in degeneracy lifting, and possible stabilization of non-collinear/flux/glassy phases.

V. **Energetics, transport, and dynamical characterization:** Using SF-MC simulations [12,13,14], compute free-energy landscapes, energy barriers, magnetization and resistivity hysteresis, Kubo–Greenwood transport [10,18], relaxation curves, and domain evolution under temperature and applied magnetic field. Identify signatures of metastability (aging, memory, slow relaxation) [3,4,19] and field-driven melting of insulating domains [5,9].

VI. **Qualitative experimental validation (Secondary Experimental Objectives):** Synthesize representative manganite samples (EPS and frustrated types) by sol–gel and solid-state techniques, and perform XRD, temperature-/field-dependent resistivity, and SQUID magnetometry to qualitatively test key simulation predictions (domain-assisted MIT shifts, hysteresis, relaxation, and field-induced switching) [2,5,6,20].

4. Research Plan and Methodology

A. Theoretical Method (Main Component)

The theoretical effort will progress through the following stages:

I. **Implementation and Validation of the Spin–Fermion Monte Carlo Framework:** The theoretical work will begin with the construction and validation of the complete spin–fermion Monte Carlo (SF–MC) model, incorporating itinerant e_g electrons, localized Mn spins, Jahn–Teller lattice distortions, superexchange interactions, and quenched disorder [1,9,10]. Initial simulations will be performed on small lattices to benchmark energy calculations, update schemes, and observables. Following successful validation, the framework will be expanded using numerical acceleration techniques such as the traveling cluster approximation [14] and polynomial expansion methods [12,13], enabling efficient simulation of larger and more realistic lattice sizes. This foundational stage ensures numerical accuracy and establishes the computational tools needed for subsequent investigation of metastable phenomena.

II. **Baseline Study of Intrinsic Phase Competition and Metastability:** After establishing the simulation framework, the next phase will involve examining phase competition and metastability in uniform bulk-like manganite models without engineered modifications [1,9,10]. These baseline simulations will characterize the natural coexistence of ferromagnetic metallic and charge-ordered insulating regions, the formation of electronic phase separation [1,5], and the emergence of metastability manifested through hysteresis, relaxation, and energy-barrier structures [3,4,19]. By mapping free-energy landscapes and tracking domain evolution under temperature and magnetic-field sweeps [9,10,11], this stage provides a reference description of intrinsic behavior against which all later structural and electronic modifications will be compared.

III. **Progressive Incorporation of Structural and Electronic Modifications:** With the baseline behavior understood, the study will systematically introduce engineered variations into the model. Ordered antidot arrays of different shapes, sizes, and periodicities will be embedded within AFM–CO backgrounds to explore how geometric constraints alter nucleation and growth of FMM domains [5,15]. Electronic filling will be varied to analyze doping-dependent changes in EPS strength, metastability, and CMR-like responses [9,10,11,16]. Simulations will also be conducted on lattices with different effective thicknesses to capture thin-film behaviors and dimensional crossover [5,6]. The progression from simple to increasingly complex modifications will help reveal how artificial structure, charge density, and geometry reshape the stability of competing phases and the pathways through which metastable states emerge [9,10,11].

IV. **Extension to Geometrically Frustrated Manganite Lattices:** The theoretical effort will further extend to geometrically frustrated lattice structures such as triangular and FCC-derived networks, where magnetic frustration naturally produces non-collinear order, flux phases, and glassy states [7,8,17]. Using the same SF–MC framework [12,13,14], these systems will be studied to determine how frustration interacts with electron–lattice coupling and disorder to generate complex metastable behavior [3,4]. Antidot patterns and boundary modifications will then be incorporated into these

frustrated geometries to evaluate how engineered structures lift degeneracies or stabilize particular metastable configurations. This stage provides insight into the combined effects of frustration and artificial structuring on the energy landscape and dynamical evolution of correlated electrons.

V. **Unified Evaluation of Physical Observables and Metastable Behavior:** At each stage of the computational work, physical observables will be computed in a consistent manner to provide a unified description of metastability and phase competition. Electronic transport properties will be obtained using the Kubo–Greenwood formalism [10,18], while magnetic behavior will be extracted from Monte Carlo–averaged spin configurations [9,10,11]. Metastability will be analyzed through thermal and magnetic-field cycling, hysteresis loops, domain evolution snapshots, and time-dependent relaxation studies [3,4,19,20]. These results will allow the construction of phase diagrams, domain maps, free-energy profiles, and dynamical signatures that jointly characterize how metastable states arise, evolve, and can be controlled through structural patterning, charge manipulation, and dimensional tuning [9,10,11].

B. Experimental Method (Supportive Component)

The experimental component of this work will serve as a qualitative validation of the theoretical predictions derived from the spin–fermion Monte Carlo simulations.

I. **Synthesis of Representative Manganite Samples:** The experimental work will focus on synthesizing a small set of manganite samples that exhibit electronic phase separation and geometrical frustration, chosen to correspond to the regimes explored in the simulations [5,6]. Samples will be prepared using the sol–gel route and conventional solid-state synthesis to ensure good phase purity, compositional control, and homogeneous microstructure [2]. These synthesis methods allow systematic adjustment of chemical composition and oxygen stoichiometry, enabling experimental conditions that reflect the theoretical model assumptions.

II. **Structural Characterization Using X-ray Diffraction:** The structural properties of the synthesized samples will be examined using X-ray diffraction (XRD) to verify phase purity, crystallographic symmetry, lattice parameters, and any strain-related distortions [2,16]. This structural information is essential for establishing correspondence with the theoretical lattice models and determining whether real samples exhibit features—such as slight distortions or secondary phases—that may influence metastable behavior. A careful structural assessment ensures that the comparison between simulation and experiment remains meaningful.

III. **Electronic and Magnetic Measurements for Qualitative Validation:** To compare the theoretical predictions with real materials, temperature- and magnetic-field–dependent resistivity measurements will be carried out using a standard four-probe method to detect metal–insulator transitions, resistive hysteresis, and relaxation effects [2,5,6,18,20]. Additionally, SQUID magnetometry will be used to study magnetic

ordering, coercivity, thermal hysteresis, memory effects, and long-time relaxation [5,6,20]. These transport and magnetic measurements will provide qualitative signatures of metastability and electronic phase separation, allowing direct comparison with simulation outputs such as domain evolution, energy barriers, and field-driven phase transitions [5,9,10].

5. Expected Outcomes

The proposed study is expected to yield a detailed microscopic understanding of how metastable energy states emerge and evolve in manganites through the interplay of double exchange, superexchange, lattice distortions, geometric confinement, and frustration. Spin–fermion Monte Carlo simulations will clarify how electronic phase separation can be deliberately manipulated using ordered antidot arrays, variable charge density, and controlled film thickness—revealing mechanisms by which FMM domains nucleate, form ring-like structures, reshape energy barriers, and shift metal–insulator transition temperatures without altering chemical composition. In frustrated lattice geometries, the work will uncover how non-collinear spin textures, flux phases, and glassy states arise naturally from geometric constraints, and how engineered structural modifications lift degeneracies or stabilize specific metastable configurations. The simulations will generate comprehensive phase diagrams and free-energy landscapes that unify EPS-driven and frustration-driven metastability. The limited experimental studies, involving XRD, resistivity, and SQUID magnetometry, are expected to reproduce key qualitative behaviors—such as resistive hysteresis, magnetic memory, slow relaxation, and field-induced melting of insulating states—thereby validating the theoretical predictions and demonstrating the physical relevance of the proposed mechanisms.

6. Significance of the Study

The proposed study will provide a unified microscopic framework for understanding and controlling metastable states in manganites—an area where experiments have advanced rapidly but theoretical interpretation remains incomplete. By integrating antidot engineering, charge-density tuning, and dimensional effects within spin–fermion Monte Carlo simulations, this work will reveal how free-energy landscapes and phase competition can be deliberately modified in correlated oxides. Such insights are significant for guiding the design of oxide devices that rely on tunable metal–insulator transitions, nonvolatile memory elements, and metastability-driven functionalities. Additionally, extending the analysis to frustrated lattices will broaden understanding of emergent non-collinear and glassy phases, contributing to the broader physics of frustrated magnetism. The limited experimental validation will ensure that the theoretical predictions remain physically grounded, enhancing their relevance for future materials research and device applications.

7. Chapterization

This PhD thesis proposal will be divided into several chapters as follows:

Chapter 1: Introduction and Literature Review

This chapter will introduce the physics of manganite systems, emphasizing the competing interactions—double exchange, superexchange, Jahn–Teller effects, and disorder—that give rise to metastable states and electronic phase separation. It will also review experimental and theoretical progress in EPS, CMR, geometrical frustration, slow relaxation, and antidot- or geometry-induced phase control. Existing simulation work and unresolved questions will be discussed to establish the motivation and necessity of the present study.

Chapter 2: Methodology

This chapter will describe the theoretical and experimental approaches used. The spin–fermion Hamiltonian, lattice geometries, boundary conditions, and numerical Monte Carlo scheme will be detailed, along with the incorporation of engineered features such as antidots, variable charge density, and thickness effects. Complementary experimental procedures for synthesizing and characterizing manganite samples using XRD, resistivity, and SQUID magnetometry will also be outlined.

Chapter 3: Results and Discussion

This chapter will present the results of the simulations and their physical interpretations. It will analyze metastable states, phase competition, free-energy landscapes, hysteresis, and relaxation behavior in uniform, antidot-engineered, and frustrated manganite systems. Comparisons with experimental findings—such as resistive hysteresis, magnetic memory, and field-induced transitions—will be made to establish consistency between theoretical predictions and observed phenomena.

Chapter 4: Conclusion

This chapter will summarize the major findings regarding engineered phase competition and metastability in manganite systems. It will synthesize insights gained from antidot arrays, charge-density variation, dimensional confinement, and frustrated lattice geometries, presenting a unified understanding of metastable behavior.

Chapter 5: Summary and Scope for Future Work

This chapter will consolidate the overall achievements of the research and discuss broader implications for correlated electron systems. It will outline future directions such as ultrafast dynamical studies, multiscale modelling, machine-learning-guided phase engineering, and potential device-oriented applications involving metastable states in manganites.

Chapter 6: References

A comprehensive list of all sources cited in the thesis, covering foundational works in manganite physics, metastability, electronic phase separation, geometric frustration, antidot engineering, and theoretical modeling.

Additional chapters may be incorporated as the research progresses, and the content of individual chapters may be modified or expanded based on the actual findings and scope of the work.

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Declaration

I hereby declare that the above synopsis titled**"Exploring Metastable Energy States through Engineered Phase Competition in Manganite Systems"**is submitted as part of the requirement for the Ph.D. Programme(PAT-2023) at the University Department of Physics, Bhupendra Narayan Mandal University (BNMU).

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